• Analog Elements for Transuranic Chemistries

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The analytical technique for measuring trace concentrations of the analog rare earth elements has been refined for optimal detection. The technique has been used to determine the rare earth concentrations in a series of geological and biological materials, including samples harvested from controlled lysimeter investigations. These studies have demonstrated that any of the trivalent rare earth elements may be used as analog elements for the trivalent transuranics, americium and curium.

BEHAVIOR OF ANALOG AND TRANSURANIC ELEMENTS IN THE ENVIRONMENT

The objective of this program is to examine the environmental behavior of selected naturally occurring analog elements and to use these observations to predict the long-term behaviors of the transuranic elements americium and curium. The predicted transuranic behavior will eventually be used to estimate soil-to-crop transfer and food-chain transport of americium and curium that have reached an environmental equilibrium condition. The ultimate objective is to estimate the dose-to-man that these radionuclides would represent. Thus far we have: evaluated the soil chemistries of the rare earth analog elements (especially the element neodynium); measured soil-to-crop transfer for several selected species; and established, planted and harvested a series of lysimeters that were amended with both analog and transuranic elements. The lysimeters are used to measure soil-to-plant transfer and evaluate the rates at which selected species approach the chemical behavior of the native analog elements.

Short-term investigations (up to several weeks for plant uptake studies) have proven that americium, curium and their natural analog elements behave identically in the study regimes. The same chemical forms of the rare earth and transuranic elements behave the same. In addition, the pool of natural rare earth analog elements that has been defined as being biologically available in soil systems behaves approximately the same as does a transuranic from another source (fallout ^{241}Am) in soil-to-plant transfer. Thus, these analog elements are good predictors of the transuranic behaviors. Study of the transport of the analog elements, although difficult, is achievable. These data also suggest that the radionuclides originating from the fallout from atmospheric testing programs are themselves good predictors of the ultimate environmental behavior of radionuclides entering the environment from other sources. However, the extremely low levels of fallout transuranic elements in the environment make the study of their transport through the food chain extremely difficult.

Investigations currently in progress include: 1) the determination of rare earth concentrations in a few selected animal species and their controlled diets in order to evaluate the plant-to-animal transfer of the analogs; 2) continued analysis of pea samples grown on lysimeters that have previously been spiked with americium, curium, cesium, and neodymium isotopes to evaluate how the uptake of these elements varies with time; and 3) selective chemical leaching of soils from some of the lysimeters in order to evaluate the comparative geochemistries of the added elements and those of the naturally occurring analogs in the soil.

RARE EARTH ELEMENT ANALYSIS BY RADIOCHEMICAL NEUTRON ACTIVATION

The concentrations of the rare earth elements in biological materials are extremely low, at the ppb to ppt level. In order to measure these concentrations precisely in plant or animal samples, a very sensitive analytical technique has been developed and revised. Briefly, this technique is a neutron activation analysis in which the sample is irradiated, then fused with nonactivated rare earth element carriers. The rare earth elements are separated from the remaining ions in the sample, and the individual elements in this enriched rare earth fraction are quantified by counting on a detector which has been selected to optimize detection of specific rare earths. After counting, the samples are reirradiated and recounted to determine the overall chemical yield for the inactive carriers, which had been added prior to the rare earth element group separation.

TABLE 1. Comparison of LEPD, Normal Ge (Li) and Ge (Li)-NaI(tl) Noncoincidence Detections for Rare Earths

Isotope	Decay[b] Interval	E (keV)	Relative Intensity[a] LEPD	Ge (Li)	Ge (Li) - NaI (Tl) Noncoincidence	Favorable Mode
^{140}La	1	329	0.36	0.61	—	
		487	0.10	1.0	—	Ge (Li)
		816	—	0.38	—	
		1596	—	0.73	—	
^{141}Ce	3	145	1.0	1.0 (13)[c]	0.86 (20)	Noncoincidence
^{143}Ce	1	293	0.53	1.0 (2.0)	0.68 (3.2)	Noncoincidence
^{142}Pr	1	1576	—	1.0 (2.0)	1.1 (5.0)	Noncoincidence
^{147}Nd	2,3	91	1.7	1.0 (2.0)	0.70 (2.0)	LEPD
		531	—	0.22 (3.5)	0.20 (10)	
^{153}Sm	1,2	41	1.8	0.14	—	LEPD
		70	0.17	0.10 (4.0)	0.055 (4.0)	
		103	1.1	1.0 (3.0)	0.74 (50)	Noncoincidence
^{152m}Eu	1	842	—	1.0 (5.5)	0.70 (10)	Ge (Li), Noncoincidence
^{152}Eu	3	122	0.92	1.0 (17)	0.20 (6.0)	LEPD, Ge (Li)
		344	0.26	0.44 (30)	0.16 (30)	
		1408	—	0.082 (50)	0.050 (50)	
^{159}Gd	1	363	0.67	1.0 (1.5)	0.65 (2.0)	Ge (Li) Noncoincidence
^{153}Gd	3	98	1.1	1.0 (2.0)	0.68 (2.2)	All
^{160}Tb	3	87	1.2	0.80	—	
		298	0.60	1.0	—	Ge (Li)
		879	—	0.42	—	
^{166}Ho	1	81	1.4	1.0 (3.0)	0.65 (3.0)	LEPD
^{177}Er	1	308	0.60	1.0 (1.6)	0.80 (3.0)	Noncoincidence
^{170}Tm	3	84	1.3	1.0 (2.8)	0.80 (4.0)	All
^{175}Yb	1,2	283	0.30	0.53 (8.0)	0.31 (12)	
		396	0.40	1.0 (24)	0.80 (50)	Noncoincidence
^{169}Yb	3	63	3.0	1.4 (5.0)	0.35 (3.0)	LEPD
		131	0.50	0.56 (4.0)	0.16 (3.0)	
		177	0.67	1.0 (8.0)	0.50 (8.0)	Ge (Li)
^{177}Lu	1,2	208	0.71	1.0 (9.0)	0.61 (11)	Ge (Li), Noncoincidence

(a) Data normalized to prominent photopeak in normal Ge (Li) spectra. The prominent gamma rays are underlined.
(b) (1) 2 to 5 days decay; (2) 7 to 11 days decay; (3) 20 to 30 days decay.
(c) The values in parentheses are peak to compton ratios in a given decay interval.

Three different counting systems and three radioactive decay periods were evaluated for quantifying the series of rare earth elements in biological samples. The counting system included: 1) a normal Ge(Li) detector (130 cm, 26% efficiency, full width at half maximum of 1.8 KeV for 1332 Key γ of ^{60}Co) at 0.50 KeV/channel; 2) a 19 cm^2 surface area low-energy photon detector (planar, intrinsic Ge; full width at half maximum of 0.71 KeV for 122 KeV γ of ^{58}Co) at 0.2 KeV/channel for x-ray and low-energy (<400 KeV) γ-rays; and 3) a coincidence-noncoincidence Ge(Li)-NaI(Tl) detector system (130 cc, 26% efficiency, full width at half maximum of 1.9 KeV for 1332 KeV γ of ^{60}Co) at 0.5 KeV/channel. For biological materials, detection of the trace rare earth element concentrations was enhanced with the low-energy photon and the coincidence-noncoincidence detectors. To compare the relative detection sensitivities of the individual rare earth elements by these three systems, we have calculated ratios of specific activities of the various rare earth isotopes in a rare earth standard that was counted on each of the counting systems. These data are presented in Table 1. The most favorable mode for detection of an individual rare earth can be determined by comparing the numerical values of these relative intensity ratios.

Based on the comparison shown in Table 1, normal Ge(Li) counting is favored for ^{140}La and ^{160}Tb; noncoincidence counting for ^{141}Ce, ^{143}Ce, ^{142}Pr, ^{153}Sm (103 KeV) ^{171}Er, and ^{175}Yb; and low-energy photon detection counting for ^{147}Nd, ^{153}Sm (41 KeV), ^{166}Ho, and ^{169}YB. The detection of ^{152m}Eu, ^{159}Gd (363 KeV), and ^{177}Lu is equally sensitive by normal Ge(Li) and non-coincidence counting; ^{152}Eu (122 KeV) detection is equally sensitive by both low energy photon detection and normal Ge(Li); and ^{153}Gd (98 KeV) and ^{170}Tm are equally sensitive by all counting modes. Since neodymium is the principle analog for americium and curium, low energy photon detection is the preferred counting mode. For those samples where the concentrations of the whole series of rare earths are desired, the samples are also counted on the coincidence-noncoincidence counting system. This approach provides optimum analytical sensitivity for nearly the entire series of rare earth elements.